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D E C I S I O N
of 26 November 1996

Case Number: T 0643/93 - 3.3.3
Application Number: 88105217.9
Publication Number: 0286005
IPC: C08G 18/50
Language of the proceedings: EN

Title of invention:
Flexible polyurea or polyurea-polyurethane foams prepared from high equivalent weight amine-terminated compounds and process for preparing the same

Applicant:
THE DOW CHEMICAL COMPANY

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (yes) - comparison with variant lying closer than relevant state of the art - no expectation of improvement"

Decisions cited:
T 0035/85

Catchword:
-



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Boards of Appeal

Chambres de recours

Case Number: T 0643/93 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 26 November 1996

Appellant:

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Decision under appeal:

Decision of the Examining Division of the
European Patent Office dated 3 March 1993
refusing European patent application
No. 88 105 217.9 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: R. Young
J. A. Stephens-Ofner

Summary of Facts and Submissions

- I. European patent application No. 88 105 217.9, entitled "Flexible polyurea or polyurea-polyurethane foams prepared from high equivalent weight amine-terminated compounds and process for preparing the same", filed on 30 March 1988 and published under No. 0 286 005, was refused by a decision of the Examining Division dated 3 March 1993, on the ground of lack of inventive step with regard to the disclosure of the document D1: US-A-3 256 213. The decision was based on a set of twelve claims, of which Claim 1 read as follows:

"An open-celled flexible polyurea or polyurea-polyurethane foam having a density of 3 lbs/cu. ft (50 kg/m³) or less, which is the reaction product of a reaction mixture comprising

- (A) a high equivalent weight compound or mixture thereof, the equivalent weight being in the range of from 400 to 5000 and having an average from 1.5 to 4 active hydrogen-containing groups per molecule, of which active hydrogen-containing groups, at least 50 percent are primary aromatic, Lewis acid-blocked primary aliphatic and/or secondary aliphatic or aromatic amine groups,
- (B) a blowing agent including at least 3.5 parts of water per 100 parts component (A)

- (C) a cross-linker having from 3 to 8 active hydrogen-containing groups per molecule and an equivalent weight of 200 or lower and/or a chain extender having two active hydrogen-containing groups per molecule and an equivalent weight of from 31 to 300, the total weight of the cross-linker and/or the chain extender being less than 5 parts by weight per 100 parts of component (A), and
- (D) a polyisocyanate."

Claims 2 to 9 were dependent claims directed to elaborations of the foam according to Claim 1.

Claim 10, an independent claim, read as follows:

"An active hydrogen-containing composition comprising

- (A) a high equivalent weight compound or mixture thereof, the equivalent weight being in the range of from 400 to 5000 having an average of from 1.5 to 4 active hydrogen-containing groups per molecule, of which active hydrogen-containing groups, at least 50 percent are primary aromatic, Lewis acid-blocked primary aliphatic and/or secondary aliphatic or aromatic amine groups,
- (B) from 3.5 to 10 parts by weight of water per 100 parts by weight of component (A), and
- (C) a cross-linker and/or chain extender, both as defined in claim 1, the total weight of the cross-linker and/or chain extender being from 0.1 to 4.5 parts by weight per 100 parts by weight of component (A)."

Claim 11 was directed to a process for preparing an open-celled polyurethane foam comprising reacting a reaction mixture as defined in Claim 1 in a one-step process.

Claim 12 was directed to an elaboration of the process of Claim 11.

According to the decision, Claim 1 of the application differed from Example 11 (second part) of D1 only by the amount of water used - this being lower in Example 11 than the minimum of 3.5% in Claim 1 of the application in suit. Such a difference was held not to be inventive, however, since no convincing evidence had been filed showing that the claimed foams had better properties than those of Example 11 in D1. Furthermore, there was a statement in D1 that higher amounts of water could be used (D1, column 5, lines 55, 56).

II. On 30 April 1993, a Notice of Appeal against the above decision was filed, together with payment of the prescribed fee.

In the Statement of Grounds of Appeal filed on 1 July 1993, the Appellant (Applicant) disagreed with the latter finding, on the basis that the whole teaching of D1 was to find a way of reducing the amount of water used, and therefore the consumption of isocyanate in the reaction, and that the reference to the possibility of using higher amounts of water was in fact a warning that this would produce a foam of worse properties (Statement of Grounds, page 2, second paragraph).

Moreover, the fact that over 20 years had elapsed between the publication of D1 and the filing of the application in suit was an indication that this step was indeed not obvious.

III. Following the issue, on 18 September 1996, of a communication by the Board accompanying a summons to oral proceedings, in which the objection was additionally raised that the subject-matter claimed in the application lacked novelty in the light of D1, the Appellant filed, on 25 October 1996, further amended versions of Claims 1 and 10.

IV. Oral proceedings were held before the Board, in accordance with the request of the Appellant, on 26 November 1996.

During the oral proceedings, the Appellant filed a new, restricted set of Claims 1 to 10 which formed the sole request relied upon. Claim 1 reads as follows:

"Use of a reaction mixture for the preparation, in a one-shot process, of an open-celled flexible polyurea or polyurea-polyurethane foam having a density of 1.5 lbs/cu. ft (25 kg/m³) or less, said reaction mixture comprising

- (A) a high equivalent weight compound or mixture thereof, the equivalent weight being in the range of from 400 to 5000 and having an average from 1.5 to 4 active hydrogen-containing groups per molecule, of which active hydrogen-containing groups, at least 50 percent are primary aromatic, Lewis acid-blocked primary aliphatic and/or secondary aliphatic or aromatic amine groups,
- (B) a blowing agent including at least 7.5 parts of water per 100 parts component (A),
- (C) a cross-linker having from 3 to 8 active hydrogen-containing groups per molecule and an equivalent weight of 200 or lower and/or a chain extender having two active hydrogen-containing groups per

molecule and an equivalent weight of from 31 to 300, the total weight of the cross-linker and/or the chain extender being less than 5 parts by weight per 100 parts of component (A), and

(D) a polyisocyanate,

wherein the polyisocyanate is present in an amount sufficient to provide an isocyanate index of from 90 to 125."

Claim 9, an independent claim, reads as follows:

"An active hydrogen-containing composition consisting of

- (A) a high equivalent weight compound or mixture thereof, the equivalent weight being in the range of from 400 to 5000 having an average from 1.5 to 4 active hydrogen-containing groups per molecule, of which active hydrogen-containing groups, at least 50 percent are primary aromatic, Lewis acid-blocked primary aliphatic and/or secondary aliphatic or aromatic amine groups,
- (B) from 7.5 to 10 parts by weight of water per 100 parts by weight of component (A), and
- (C) a cross-linker and/or chain extender, both as defined in claim 1, the total weight of the cross-linker and/or chain extender being from 0.1 to 4.5 parts by weight per 100 parts by weight of component (A)."

Claims 2 to 8 and 10 are directed to elaborations of the use according to Claim 1.

- V. The Appellant requested that the decision under appeal be set aside, and a patent granted on the basis of the set of claims filed at the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.
2. *Amendments*

According to the decision under appeal, the claims then under consideration were admissible in the sense of Article 123(2) EPC (Reasons for the decision, point 2). The Board sees no reason to take a different view.

- 2.1 Claim 1 differs from that version in three respects:

- (a) The lower limit of the amount of water in Component (B), the blowing agent, has been raised to at least 7.5 parts of water, per 100 parts of component (A).
- (b) The claim is now directed to the use of a reaction mixture for the preparation, in a one shot process, of an open-celled flexible polyurea or polyurea-polyurethane foam having a density of 1.5 lbs/cu. ft (25 kg/m^3) or less, rather than to an open-celled foam per se, having a density of 3 lbs/cu. ft (50 kg/m^3) or less;
- (c) The quantity of isocyanate in the composition is required to provide an isocyanate index of 90 to 125.

Claim 9, which corresponds to Claim 10 underlying the decision under appeal, has been restricted in the following two respects compared with the corresponding previous version:

- (i) The lower limit on the amount of water forming component (B) is now required to be at least 7.5 parts by weight of water per 100 parts of component (A).
- (ii) The composition is defined as consisting of, rather than comprising, the components (A), (B) and (C).

Of the remaining claims, Claims 2 to 8, which are dependent on Claim 1, correspond to Claims 3 to 9, and Claim 10 to Claim 12 of the previous version, respectively. Previous Claim 11 has been deleted.

- 2.2 Amendments (a) and (i) are supported by the description as originally filed on page 15, lines 34 to 37 (published application, page 6, lines 16 to 17), relating to the range above 7.5 parts of water, when read in conjunction with Example 2, Sample No. 7, as originally filed, relating to the lower limiting value of 7.5 parts of water. Amendment (b) is supported by the previous text of the claim, since the same reaction mixture is involved in each case, read in conjunction with the description as originally filed on page 19, lines 18 to 21 (published application, page 7, lines 21, 22) in which it is made clear that it is essential that the process is a "one-shot" process; and by the description, as originally filed, on page 15, lines 43 to 45 (published application, page 6, lines 19

to 21), wherein a foam density of less than 1.5 pounds per cubic foot is referred to in connection with the amounts of water to which the claim has been limited by amendment (a). Amendment (c) is supported by Claim 2 as originally filed.

The basis for amendment (ii) is the previous text of the claim, since the latter clearly discloses a composition consisting of the relevant components.

2.3 In view of the above, the amendments effected do not give rise to objection under Article 123(2) EPC.

3. *The closest state of the art; the technical problem*

The application in suit is concerned with the production of an open-celled flexible polyurea or polyurea-polyurethane foam which has good load-bearing properties, in which the use of cross-linkers or microdispersions of polymeric filler materials, can be minimised or even eliminated, and to compositions capable of reacting, in a one-step process to yield such a foam. The process involves the use of a reaction mixture containing, as essential components, (A) a high equivalent weight compound or mixture thereof, the equivalent weight being in the range of from 400 to 5000 and having an average of from 1.5 to 4 active hydrogen-containing groups per molecule, at least 50 percent of which are primary aromatic, Lewis acid-blocked primary aliphatic and/or secondary aliphatic or aromatic amine groups, and (B) a blowing agent including water (published application, page 2, lines 46 to 48; page 3, lines 9 to 38).

Such compositions are, however, known from D1, which is the closest state of the art.

3.1 According to D1, cellular isocyanate reaction products having excellent load-bearing properties and wherein the amount of water and of polyisocyanate required are substantially reduced, are produced from compositions having long chain polyamino organic compounds as one of the principal components reactive with isocyanate. The compounds, which have a molecular weight of 500 to 6 000, are free of groups other than terminal amine groups which react with isocyanato groups (column 1, lines 11 to 17; 40 to 45; 50 to 52, and 58 to 72). They may, however, be mixed with a polyalkylene ether polyol also having a molecular weight of from 500 to 6 000, and a blowing agent such as a fluorocarbon or carbon dioxide (column 2, lines 17 to 36). Small amounts of water may be added to facilitate blowing. In general, it is preferred that water is employed in amounts of less than two parts by weight per 100 parts by weight of the combined polyamino compound and polyalkylene polyol. Higher amounts of water can, of course, be employed. However, no commensurate advantages are obtained thereby (column 5, lines 47 to 58).

Example 11 (first part) describes the preparation of a flexible polyether-polyurethane type foam from the following composition:

Ingredients:	parts by weight
Union Carbide LG56 triol (a polypropylene oxide adduct of glycerol, hydroxyl No. 56).....	75
p-diamino benzoate ester of polybutadiene glycol..	25
Polysiloxane-oxyalkylene block polymer.....	1.0
Stannous octoate.....	0.3
Dichloromonofluoromethane.....	10.0
N-methyl morpholine.....	0.25
N,N,N',N'-tetramethylbutanediamine.....	0.03
Water.....	1.72
80/20 isomer ratio 2,4/2,6-toluene diisocyanate...	24.50

There resulted a foam having inter alia a density of 1.35 lb/cu.ft.

In the second part of the example, a similar foam was made using 50 parts of the polybutadiene diamine and reducing the propylene triol (LG 56) to 50 parts. This foam had a density of 1.45 lb/cu.ft.

- 3.2 According to the decision under appeal, the disclosure of D1, Example 11, second part, differed from the subject-matter claimed in the application in suit only by the use of lower amounts of water (Section I., last paragraph, above).
- 3.3 Compared with this state of the art, the technical problem underlying the application in suit may be seen as the search for an ultra-low density foam having improved load-bearing properties.
- 3.4 The solution proposed according to the application in suit is to increase the amount of water used as blowing agent to at least 7.5 parts by weight per 100 parts by weight of the isocyanate-reactive component, i.e. of the component consisting of the long chain polyamino compound and any admixed polyalkylene ether polyol.
- 3.5 A direct comparison of the relevant load-bearing properties (tensile strength, tear strength, and resiliency) involving the foams prepared according to Example 11 of D1 is not possible, since no values are given in relation to this example, and no evidence in this respect was filed by the Appellant, despite repeated invitations of the Examining Division to do so (cf. Decision under appeal, Reasons, page 6, first two paragraphs).

3.6 Nevertheless, evidence is available from the application in suit itself which is, in the Board's view, relevant, at least in relation to compositions having an amount of water corresponding to the new, raised lower limit to which the claims of the application in suit have been limited.

3.6.1 According to the results (Table IV) of tests performed on foams prepared according to Example 2 of the application in suit, in which the reactive mixtures differed essentially only in the quantity of water used (cf. Table III), it can be seen that, compared with a foam made using a lower quantity of water as blowing agent (Sample No. 3; 3.8 parts of water per 100 parts of amine-terminated polyether), a foam made using 7.5 parts of water per 100 parts of amine terminated polyether (Sample No. 7) had markedly improved values of tensile strength (0.13 MPa instead of 0.10 MPa), tear strength (308 N/m instead of 207 N/m) and resiliency (46% instead of 50%). These improved mechanical properties are obtained despite the fact that the foam has a considerably lower density (19 kg/m³ rather than 32 kg/m³).

3.6.2 The above comparison Sample No. 3 represents a variant lying closer to the claimed subject-matter than the closest state of the art represented by D1, Example 11. It has been held by another Board, that the Applicant or Patentee may discharge his onus of proof by voluntarily submitting comparative tests with newly prepared variants of the closest state of the art making identical the features common with the invention (T 0035/85 of 16 December 1986, not published in OJ EPO). The comparison in the application in suit referred to above is regarded as such a voluntary comparative test, and consequently as a fair comparison.

3.6.3 . Furthermore, taken in conjunction with the general statement in the description of the application as filed (page 15, second paragraph; published application, page 6, lines 16, 17) that "With this invention, in excess of 7.5 parts of water can be used per 100 parts of amine-terminated compound to provide an ultra-low density foam having useful physical properties", which the Board has no reason to doubt, it is credible that the measures proposed provide an effective solution of the stated problem.

4. *Novelty*

There is no disclosure of a composition in which water is present in an amount of 7.5 parts or more per 100 parts by weight of isocyanate-reactive component, which is now an essential feature of both independent Claims 1 and 9.

Consequently, the claimed subject-matter is novel.

5. *Inventive step*

It is necessary to consider whether the person skilled in the art, starting out from the foams of D1, would have expected improved load-bearing properties, and in particular higher tensile strength and tear strength as well as lower resiliency (i.e. greater stiffness), at a comparable level of density, would result from increasing the amount of water in the blowing agent to at least 7.5 parts per 100 parts by weight of the combined polyamino organic compound and the polyalkylene ether polyol.

5.1 To effect this modification in the teaching of D1 would involve increasing the water content in the relevant Example 11 (second part) which is 1.72 parts per 100 parts by weight of the combined polyamines organic compound and the polyalkylene ether polyol (section 3.1, last three paragraphs, above) by about a factor of four, and, compared with the maximum amount of water mentioned in the general description of D1 (section 3.1, first paragraph, above), a factor of well over three.

The required amount of water is also almost double the maximum acknowledged in the application in suit as being "conventional" in the production of polyol-based polyurethanes (2-4 parts per 100 parts by weight of polyol), and considerably more than the maximum, of 4.6 parts, acknowledged in the application as having been previously achieved in the art (application as filed, page 3, final paragraph; published application, page 2, lines 41 to 43).

Whilst it is true that the disclosure of D1 opens up the possibility of using larger amounts of water, it is stated that no commensurate advantages are obtained thereby (column 5, lines 55 to 58).

Consequently, the skilled person would have no expectation from the teaching of D1 that an improvement in the relevant load-bearing properties would result from such an increase in the amount of water used in the blowing agent. On the contrary, the expected effect of increasing the amount of water would be to consume isocyanate and decrease the density of the resulting foam, which would in turn weaken it.

In other words, the solution provided is not obvious from the disclosure of D1.

5.2 Hence, the subject-matter of Claims 1 and 9 involves an inventive step within the meaning of Article 56 EPC. By the same token, the subject-matter of Claims 2 to 8 and 10, which are dependent on Claim 1 also involves an inventive step.

Order

for these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the Examining Division with the order to grant a patent with the claims submitted in the course of oral proceedings as the sole request and after corresponding amendments of the description.

The Registrar:



E. Gorgmaier

The Chairman:



C. Gérardin